

U.S. Application Serial No.: 10/699,258  
Atty. Docket No.: JCW-0306  
Reply to Office Action of August 22, 2006

### REMARKS / ARGUMENTS

In response to Examiner's Office communication dated August 22, 2006, applicants offer the following responses.

### CLAIM OBJECTIONS

The Examiner has objected to the use of the expression "a SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratio" in line 8 of claim 1 stating that it is unclear if this is a mass ratio or a molar ratio.

#### Applicant's Position

The term "SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratio" as utilized in the present application is in terms of a molar ratio. It is applicants' position that the term "SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub>" ratio as used in the present application is used only in context with the SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratio of a zeolite and in this context, it is clear to one of ordinary skill in the art that the SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratios identified are in molar ratios.

SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> mass ratio values in and of themselves are not important to one of ordinary skill in the art when analyzing zeolitic structures. What is directly important in zeolite structure analysis is the molar ratio values of the crystalline components in determining the structure characteristics including the acidity level of the corresponding zeolite and the amount of offsetting cations required for a balanced charge in the zeolite structure.

Secondly, it can be seen that the terminology of interchanging the terms "SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> molar ratio" and "SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratio" in the art is common practice to those of skill in the art. This practice can be seen for instance in the Olson patent (U.S. Patent 6,488,741) that was cited in the current office action. Olson refers to the Si:Al in terms of a molar ratio (see Abstract and column 2, line 61) while at other times referring to the ratio only as "Si:Al ratio" (see column 5, line 5), "silica to alumina ratio" (see column 11, line 40) or simply "SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratios" (see column 5, line 34).

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It can similarly be seen that it is common practice in the art to cite the  $\text{SiO}_2/\text{AlO}_3$  molar ratios in the shorthand term "SiO<sub>2</sub>/AlO<sub>3</sub> ratio" in view of the Eberly et al. (U.S. Patent 3,591,488). For example, in Eberly, Figure 4 is described as showing "the effect of silica to alumina molar ratios on the unit cell size of the zeolite" (column 4, lines 57-58). However, the title of Figure 4 of Eberly is the "calculated effect of SiO<sub>2</sub>/AlO<sub>3</sub> ratio on unit cell size". Here, it can again be seen that to those skilled in the art " SiO<sub>2</sub>/AlO<sub>3</sub> molar ratio" and "SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratio" are used interchangeably.

Additionally, an affidavit is enclosed by applicants from one of the current inventors confirming that the usage of both terms is commonly used in the art and that it is well understood by those of ordinary skill in the art that the short-hand term "SiO<sub>2</sub>/AlO<sub>3</sub> ratio" as used herein is synonymous to molar ratios when used within the context of analyzing zeolite compositions.

Therefore, applicants respectfully request the Examiner to reconsider and withdraw this objection.

#### REJECTION UNDER 35 U.S.C. 112

The Examiner has rejected claim 2 under 35 U.S.C. 112, second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter which applicant regards as the invention.

#### Examiner's Position

The Examiner has rejected claim 2 under 35 U.S.C. 112, second paragraph, stating that the use of the word "type" in the claims renders the claim indefinite because it is unclear what CHA adsorbent is to be used in the process.

#### Applicant's Position

It is applicants' position that the use of the term "type" in the original claims does not render the claimed invention indefinite. Nonetheless, applicants have

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amended the claims to remove the term "type" as the scope of the claim is not altered by this amendment.

It is common practice to refer to zeolite framework structures, such as the CHA framework structure, as "types" which would be synonymous in the art to the term "class" or "classification" which is clear and unambiguous. However, the use of the term "CHA structure" in lieu of the term "CHA structure type" in claim 2 does not alter the scope and meaning of the claim. Therefore, applicants have amended claim 2 to eliminate the term "type" from the claims.

Therefore, applicants respectfully request the Examiner to reconsider and withdraw this rejection.

#### **FIRST REJECTION UNDER 35 U.S.C. 103**

The Examiner has rejected claims 1-3 and 7-9 under 35 U.S.C. 103(a) as being unpatentable over Olson (6,488,741).

#### **Examiner's Position**

It is the Examiner's position that Olson discloses a process of selectively adsorbing propylene in a mixture of propylene/propane through the use of zeolites having structures of a maximum of 8-member tetrahedral rings controlling the diffusion rate. Zeolites are those of the CHA (e.g., SSZ-13) and ITE structures.

It is Examiner's position that although Olson does not disclose a zeolite with a silica to alumina molar ratio of greater than 50 but less than 200, that it would have been obvious for one to use a zeolite of less than 200.

#### **Applicant's Position**

It is applicants' position that the claims 1-3 and 7-9 are not obvious in light of Olson. Olson specifically limits the disclosure and claims of the patent to silica to alumina ratios of at least 200 (see Olson, column 2, line 60-61 and Claim 1). A problem that exists with low silica to alumina ratio zeolites is the fact that at low

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SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratios (e.g., below 200), very adverse process affects are seen. This can be seen in Figure 1 of the present application where a CHA zeolite (here H-SSZ-13) at low SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratios (SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratio = 20 in Figure 1) results in severe polymerization of the propylene resulting in almost no recovery (i.e., desorption) of the desired propylene product.

In fact, it can be seen in the examples of Olson that zeolites of only extremely high SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratios were tested. In Examples 1-10 only pure silica zeolites (SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> = infinity) were utilized in the adsorption tests. In the remaining Examples 11 and 12, an HZSM-5 zeolite with a SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratio of 3,300 was utilized in the adsorption testing of the examples. What is not obvious from Olson is that at low Si/Al ratios, deleterious polymerization effects are encountered.

It is can be seen from the present application that the problem of excessive polymerization has been observed at low SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> (as shown in Figure 1 of the current application) and the problem has been solved in the presently claimed invention by modifying the zeolite through ion-exchanging hydrogen form of the zeolite with an alkali metal cation to significantly minimize the amount of product polymerization and significantly improve the recovery rate of propylene (comparing Figures 1 and 2 of the present application).

Claims 1-3 and 7-9 require the use of "alkali metal cations" in the zeolite for which it has been discovered that propylene recovery can be significantly increased while allowing the use of a low SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratio zeolite. The discovery of the use of ion-exchanged alkali metal cations in a zeolite with a SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratio greater than 50 and less than 200 to significantly improve the recovery of the adsorbed compound as discovered in the presently claimed invention is not taught by Olson.

Therefore, the Examiner is respectfully requested to reconsider and withdraw the 35 U.S.C. 103 rejection of claims 1-3 and 7-9.

**SECOND REJECTION UNDER 35 U.S.C. 103**

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The Examiner has rejected claims 4-6 under 35 U.S.C. 103(a) as being unpatentable over Olson (6,488,741) as applied to claim 1 above, and further in view of either Eberly et al. (3,591,488) or Wakita et al. (6,579,347).

**Examiner's Position**

It is the Examiner's position that Olson discloses a process of selectively adsorbing propylene in a mixture of propylene/propane through the use of zeolites having structures of a maximum of 8-member tetrahedral rings controlling the diffusion rate. Zeolites are those of the CHA (e.g., SSZ-13) and ITE structures, but that Olson does not specifically disclose a step for dealuminating the zcolite by using steam. It is Examiner's position that both Eberly and Wakita teach a step of dealuminating a catalyst by steam and therefore it would have been obvious to one of ordinary skill in the art to have modified the Olson process by dealuminating the zcolite by steam as taught by either Eberly or Wakita.

**Applicant's Position**

It is applicants' position that the claims 4-6 are not obvious in light of Olson in view of either Eberly or Wakita. Eberly specifically states that the steaming process is performed on a zcolite to achieve greater stability to heat, steam and acid (column 2, lines 43-44). The problem solved by claims 4-6 of the presently claimed invention is to improve the adsorption/desorption properties of the zcolite. Eberly does not teach the steaming of a zcolite to improve adsorption/desorption characteristics of the zeolite. One of ordinary skill in the art with the knowledge of Olson and Eberly would not be motivated to utilize the steaming process as disclosed by Eberly to arrive at the adsorption-based process as presently claimed. Additionally, Eberly fails to provide any teachings for using alkali metal cations in the zeolite framework as per the first argument above which is deficient from the Olson reference.

Similarly, it would not be obvious to one of ordinary skill in the art in view of the Olson and Wakita references to arrive at the presently claimed invention. Firstly, Wakita teaches an adsorption process utilizing 10-ring and 12-ring zeolites (faujasites,  $\beta$ , L and MFI type zeolites, see Wakita, column 3, lines 8-11), not the 8-ring tetrahedral zeolites as presently claimed. Secondly, Wakita teaches a process for

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removal of sulfur compounds from city gas (column 3, lines 4-7) and there would be no motivation to use the steaming process which is found only in Example 6 of the Wakita reference to reduce polymerization of propylene as discovered in the present invention. Thirdly, Wakita teaches that it is "preferable that the zeolite of the present invention is treated for dealumination". This statement is in conflict with the notion that one of ordinary skill in the art and knowledge of Wakita would be motivated to steam the zeolite of the present invention if the emphasis of Wakita is to treat the zeolite to prevent alumina loss.

Finally, Wakita discloses a non-kinetic process for sulfur adsorption, not a kinetic-based propylene adsorption/desorption process as per the present invention. It can be seen that the process of Wakita is a strong equilibrium-based adsorption process and would require regeneration of the zeolite by gas sweeping, thermal heating, and/or vacuum treating (see Abstract and column 4, lines 34-41). There would be no motivation to use the teachings of Wakita in the kinetic-based process of the present invention. Additionally, Wakita fails to provide any teachings for using alkali metal cations in a low SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratio zeolite framework to improve recovery as presently claimed.

Therefore, the Examiner is respectfully requested to reconsider and withdraw the 35 U.S.C. 103 rejection of claims 4-6.

### THIRD REJECTION UNDER 35 U.S.C. 103

The Examiner has rejected claims 1-3 and 7-9 under 35 U.S.C. 103(a) as being unpatentable over Ramachandran et al. (EP 0572239 A1) in view of Addiego (EP 0768111 A1).

#### Examiner's Position

It is the Examiner's position that Ramachandran discloses separating propylene from a mixture containing propane and propylene by using an adsorbent

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(e.g. 4A zeolite) having 8-membered tetrahedral rings. The adsorbent comprises alkali metal cations (e.g., sodium).

Ramachandran does not disclose a zeolite with silica to alumina molar ratio of greater than 50 but less than 200, does not disclose CHA or SSZ-13, and does not disclose that the cations are introduced by ion exchange at a pH greater than about 7.5.

It is Examiner's position that Addiego discloses an adsorbent with a  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio of from about 50 to 250 and that it would have been obvious to one of ordinary skill in the art to have modified the Ramachandran process using the  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio disclosed in Addiego.

The Examiner also states that it would have been obvious to have modified the Ramachandran process with a zeolite such as SSZ-13 because 4A zeolite has 8-member rings of tetrahedra. In addition, the Examiner is of the position that one of ordinary skill in the art would have modified the process of Ramachandran by introducing ion exchange as a pH greater than 7.5 because it is within the level of one of ordinary skill in the art to process the zeolite at the claimed pH.

#### Applicant's Position

It is applicants' position that the claims 1-3 and 7-9 are not obvious in light of Ramachandran in view of Addiego.

Ramachandran does not disclose nor teach a kinetic adsorption process as presently claimed nor does Ramachandran teach any specific benefits of utilizing an alkali metal cation in the framework to improve adsorption/desorption properties of a low Si/Al ratio zeolite as presently claimed. These deficiencies are not overcome by the Addiego reference.

In Ramachandran, the process disclosed is not a kinetic separation process as presently claimed. This can be seen in Examples 1-8 of Ramachandran where the first step of the adsorption cycle consists of "bed equalization" (page 7, lines 12-13). A kinetic separation process as presently claimed relies on significant differences in

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the diffusion rates of the two components to be separated. A process using a specific zeolite structure in a kinetic-based process such as the present invention may not work in an equilibrium-based process such as disclosed by Ramachandran. The opposite scenario also applies. As explained above Ramachandran discloses an equilibrium-based separations process which is supported by the disclosures in the examples. Also, Ramachandran does not disclose nor teach a kinetic-based process and there is no mention of diffusion rate properties of the material that would lead one of ordinary skill in the art to use the zeolite of Ramachandran in a kinetic-based process as presently claimed.

In addition, there is no teaching or motivation by Ramachandran to ion exchange an alkali metal cation into the zeolitic framework in order to improve the adsorption/desorption properties of an 8-membered zeolite with a low SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratio zeolite (greater than 50 and less than 200) as presently claimed. Although Ramachandran discloses an alkali metal cation (e.g., sodium), there is no clear benefit understood as to the modification of this zeolite property. In fact, Ramachandran states that it may often be beneficial to exchange (i.e., remove) some of the sodium (alkali metal) from the zeolite structure and replace with other metal cations (page 4, line 18). This teaches away from any perceived benefit of having the alkali metal cation in the framework as presently claimed.

In fact, zeolite 4A is a standard material that has a SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratio equal to 1 (or very close to 1) and the Na cations are part of the synthesis process and not incorporated in post-synthesis treatment. It is known to one of ordinary skill in the art that zeolite 4A cannot be dealuminated to SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratios greater than about 10 without destroying the structure. Therefore, Ramachandran would not teach nor motivate one of ordinary skill in the art to ion exchange a alkali metal cation in a zeolite with a SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratio greater than 50 and less than 200 as such a process would destroy the structure of the 4A zeolite enabling its use as presently claimed.

Additionally, although Ramachandran states that it "may be desirable" to exchange some of the sodium ions by other metal ions (page 4, line 18), it is not clear

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what properties the "desirable results" obtain while providing a laundry list of possible ion exchanged materials containing both alkali metals and non-alkali metals wherein Ramachandran states the use of cations, "among others, potassium, calcium, magnesium, strontium, zinc, cobalt, silver, copper, manganese, cadmium, etc." (page 4, lines 23-24). Here, there is no specific teaching or motivation to select an alkali metal cation for ion exchange into a low  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio zeolite (greater than 50 to less than 200) which is not disclosed by Ramachandran to improve a kinetic based process as presently claimed.

The mere mention in the Addiego reference of a zeolite with a  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio from 50 to 250 is not motivation to combine the references as presently claimed. There is no indication, motivation, or teaching that the alkali metal cations added to a 50 to 200  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio zeolite would be beneficial to kinetic-based separations processes as presently claimed. One of ordinary skill in the art would not be motivated to combine these references to arrive at the presently claimed invention.

Additionally, neither of the references teaches or suggests the CHA structure type of claim 2 or the SSZ-13 zeolite of claim 3. Applicants are also of the position that the Examiner has provided no evidence to support his argument that one of ordinary skill in the art would have modified the Ramachandran process by introducing ion exchange at a pH of greater than about 7.5. This rejection of claim 7 is unsubstantiated by the Examiner.

Therefore, the Examiner is respectfully requested to reconsider and withdraw the 35 U.S.C. 103 rejection of claims 1-3 and 7-9.

#### **FOURTH REJECTION UNDER 35 U.S.C. 103**

The Examiner has rejected claims 4-6 under 35 U.S.C. 103(a) as being unpatentable over Ramachandran et al. (EP 0572239 A1) and in view of Addiego (EP 0768111 A1) as in claim 1 above and further in view of either Eberly et al. (3,591,488) or Wakita et al. (6,579,347).

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**Examiner's Position**

It is the Examiner's position that Ramachandran and Addiego do not specifically disclose a step for dealuminating the zeolite by using steam. It is Examiner's position that both Eberly and Wakita teach a step of dealuminating a catalyst by steam and therefore it would have been obvious to one of ordinary skill in the art to have modified the combined processes of Ramachandran and Addiego by dealuminating the zeolite by steam as taught by either Eberly or Wakita.

**Applicant's Position**

It is applicants' position that the claims 4-6 are not obvious in light of Ramachandran and Addiego in further view of either Eberly or Wakita. As stated in the argument presented above in the "Second Rejection under 35 U.S.C. 103", Eberly specifically states that the steaming process is performed on a zeolite to achieve greater stability to heat, steam and acid (column 2, lines 43-44). The problem solved by claims 4-6 of the presently claimed invention is to improve the adsorption/desorption properties of the zeolite. Eberly does not teach the steaming of a zeolite to improve adsorption/desorption characteristics of the zeolite. One of ordinary skill in the art with the knowledge of Ramachandran and Addiego and Eberly would not be motivated to utilize the steaming process as disclosed by Eberly to arrive at the adsorption-based process as presently claimed.

Similarly, it would not be obvious to one of ordinary skill in the art in view of the Ramachandran and Addiego in further view of Wakita to arrive at the presently claimed invention. Firstly, as discussed prior, Wakita teaches an adsorption process utilizing 10-ring and 12-ring zeolites (faujasites,  $\beta$ , L and MFI type zeolites, see Wakita, column 3, lines 8-11) not the 8-ring tetrahedral zeolites as presently claimed. The use of an 8-ring material is of critical importance to the present invention, especially in the separation of propylene and propane, because the kinetic-based separation process as presently claimed relies on the diffusional differentiation of the first and second components in virtue of the tailoring of the window size. In particular, when the current invention is utilized to separate propylene from propane, the propylene diffusion rate is significantly greater than the propane diffusion rate

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since the propane is significantly hindered from entering the pore structure as compared to the smaller propylene molecule. The 10-ring and 12-ring zeolites disclosed by Wakita possess pore diameters that are too large to have the kinetic-based diffusional differences as discovered in the present invention and thus the disclosures of Wakita would not lead one of ordinary skill in the art to the presently claimed invention.

Secondly, Wakita teaches a process for removal of sulfur compounds from city gas (column 3, lines 4-7) and there would be no motivation to use the steaming process which is found only in Example 6 of the Wakita reference to reduce polymerization of propylene as discovered in the present invention.

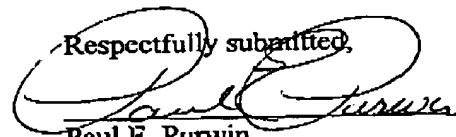
Thirdly, Wakita teaches that it is "preferable that the zeolite of the present invention is treated for dealumination". This statement is in conflict with the notion that one of ordinary in view of Wakita would be motivated to steam the zeolite of the present invention if the emphasis of Wakita is to treat the zeolite to prevent alumina loss.

Finally, Wakita discloses a non-kinetic process for sulfur adsorption, not a kinetic-based propylene adsorption/desorption process as per the present invention. It can be seen that the process of Wakita is a strong equilibrium-based adsorption process and would require regeneration of the zeolite by gas sweeping, thermal heating, and/or vacuum treating (see Abstract and column 4, lines 34-41). There would be no motivation to use the teachings of Wakita in the kinetic-based process of the present invention.

Therefore, the Examiner is respectfully requested to reconsider and withdraw the 35 U.S.C. 103 rejection of claims 4-6.

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In view of the foregoing remarks and the current amendments of the claims as presented, applicants respectfully request that the Examiner reconsider and withdraw the present claim objections and rejections and pass the case to issue.

Respectfully submitted,  


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Pursuant to 37 CFR 1.34(a)

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